Coexistence of Coulomb blockade and zero bias anomaly in a strongly coupled quantum dot

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(Dated: July 27, 2010)

The current-voltage characteristics through a metallic quantum dot which is well coupled to a metallic lead are measured. It is shown that the I-V curves are composed of two contributions. One is a suppression of the tunneling conductivity at the Fermi level and the second is an oscillating feature which shifts with gate voltage. The results indicate that Zero-Bias-Anomaly and Coulomb Blockade phenomena coexist in an asymmetric strongly coupled quantum dot.

PACS numbers: 73.63.Kv; 73.23.Hk; 73.40.Gk

Electron-electron interactions have a dramatic influence on the electronic properties of low dimensional systems. Since the dawn of solid state physics it is customary to identify two distinct contribution of interactions to electronic properties (such as the electron self energy) in solids: the Hartree and exchange terms. While the Hartree term represents the contribution of the classical Coulomb interaction, the exchange term correspond to much subtler quantum contributions.

In the tunneling conductance these two terms manifest themselves in two distinct ways: The suppression of the tunneling density of states at the Fermi level, known as the zero bias anomaly (ZBA), and the Coulomb gap/blockade. For example, tunneling into low-dimensional weakly disordered metals exhibit a pronounced ZBA phenomenon in two dimensional disordered metallic films [1] and 1D wires [2], while the Coulomb gap phenomenon dominates strongly disordered systems [3]. Experimentally, both the ZBA and Coulomb gap manifest themselves as a sharp dip in the tunneling conductivity at low source-drain voltage, V_{SD}, though the two effects have a different functional form. Since both features are centered on the Fermi energy ($V_{SD} = 0$) it is very hard to separate them.

Both terms influence also the tunneling into a quantum dot which is coupled to leads. For strongly coupled dots one expects the exchange term to dominate, resulting in a ZBA. For weakly coupled dots, the Hartree term prevents tunneling conductivity except at the degeneracy point. This leads to a zero conductance plateau in the low V regime of the I-V characteristic known as the Coulomb blockade, CB. In the case of an asymmetrical quantum dot, which is coupled more strongly to one of the leads than the other, the I-V curve exhibits a series of differential-conductance plateaus termed the Coulomb staircase (CS). Unlike the ZBA feature in which the conductance minimum is pinned to the Fermi level, the CS is sensitive to the chemical potential of the dot and oscillates with the gate voltage V_g [4]. This provides a clear way to distinguish between CB and ZBA experimentally.

For most coupling strength one of these effects will dominate over the other. For a weakly coupled dot ($g \ll 1$ where $g = hG/e^2$, G corresponding to the conductance of the dot-lead system), Coulomb blockade completely suppresses tunneling at small voltages, thus overshadowing any other e-e contribution. For tunneling into an asymmetrical strongly coupled dot ($g \gg 1$) the Coulomb blockade vanishes and the I-V curve should exhibit only a ZBA feature. What happens between these two regions, i.e., for $g \geq 1$? Can both effects be separated in the intermediate coupling regime? In this Letter we shall experimentally address these questions. Since the CB amplitude is predicted to decrease exponentially with g [5, 6], there can not be a wide coupling regime in which CB has not yet completely vanished while it is suppressed enough to allow a measurable ZBA and the two phenomena can coexist. A theoretical answer to these questions is given in a paper by Golubev *et al.* [7] who considered the I-V characteristics of an open quantum dot characterized by resistances R_S and R_D between the dot and the source/drain electrodes respectively. The electric current through the dot was found to be [8]:

$$I(V) = G_{as}V - I_0(T,V) - \tilde{G}e^{-F(T,V)}V\cos 2\pi N\,. \quad (1)$$

Here the first term describes an Ohmic current, characterized by linear conductance $G_{as} = 1/(R_S + R_D)$; the second term $[I_0(T,V)]$ reflects a conductance dip at $V = 0$ which we argue corresponds to a ZBA. The last term in Eq. (1) describes a residual CB, i.e. a part of the current that periodically oscillates with average number of electrons in the dot

$$N = \frac{C_S R_S - C_D R_D}{e(R_S + R_D)}V_{SD} + \frac{C_g}{e}V_g\,. \quad (2)$$

In this Letter we present I-V characteristics of a dot which is strongly coupled to a lead. By applying a unique method we are able to control this coupling in the regime of interest ($g \sim O(1)$). We find that the curves are composed of two parts, corresponding to two of the terms in Eq. (1) one which is oscillatory with the source-drain voltage, V_{SD}, and shifts with the application of gate voltage and the other, a conductance dip centered at $V_{SD} = 0$

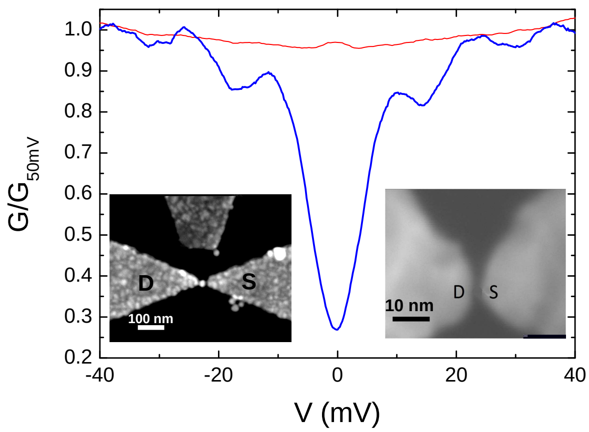

FIG. 1: Normalized differential conductance as a function of source drain voltage for tunneling directly between two gold electrodes (red light solid line) and tunneling through a gold particle (heavy solid blue line). In both cases the tunneling resistance at large voltage is $1.2M\Omega$. T=4.2K. The insets show scanning tunneling microscope images of both samples. Left inset: A 30nm gold particle strongly coupled to the drain (left) electrode and weakly coupled to the source (right) electrode. A gate electrode is fabricated at a distance of 150nm. Right inset: Two gold electrodes placed a few nm apart allowing direct source-drain tunneling.

which is V_g independent. The oscillatory feature is dramatically suppressed as the dot is increasingly coupled to the lead or as the temperature is raised. These results are interpreted as a superposition of electron interaction induced DOS suppression and the classic CS simultaneously present in a strongly coupled 0D system such that both ZBA and CB contribute to the same measurement.

The quantum dots used in this work were Au nanoparticles, 30nm in diameter. Coupling to a set of leads is performed in the following way [11, 12]: On a Si-SiO substrate we fabricate two gold electrodes (source and drain) separated by a gap of 10-30nm and a perpendicular side gate electrode at a distance of 150nm as shown in the left inset of Fig. 1. We then deposit an adhesive layer of Poly-L-Lysine on the substrate and spread gold colloids on top. Next, we use Atomic Force Microscope (AFM) nano-manipulation to "push" a desired colloid to the gap between the source and drain electrodes. This yields a quantum dot which is better connected to one of the leads than the other. We vary the coupling of the dot to the lead by depositing gold atoms on top of the electrode using an electrodeposition method. The substrate is placed in a solution containing potassium cyanaurate, potassium bicarbonate and potassium hydroxide [13]. A deposition current of $1\mu A$ is applied between the drain electrode and a gold counter electrode placed in the solution. This results in an extremely fine controllable atomic gold growth on the electrode. For each stage of the growth we cool the system to T=4.2K and measure I-V characteristics at different gate voltages.

Since the dot is better coupled to the left electrode (the drain), one can expect $R_D \ll R_S$, R_D and R_S corresponding to the resistance between the dot and the drain and the source electrodes respectively. Thus, the measured resistance through the dot $R = 1.2M\Omega$ reflects R_S. On the other hand, both the ZBA and CB effects are governed by the lower resistance, R_D, which is not directly measurable by the conductance.

Fig. 1 depicts the tunneling conductance between the source and the drain in two different cases: in the presence and absence of an Au dot connected to the drain. It is seen that in the absence of a dot, the tunneling conductance is nearly ohmic with a very small ZBA signature. This is expected since the electrodes are relatively clean (resulting in large conductance). When a dot is introduced and strongly coupled to the drain, the tunneling curves change drastically as seen in the heavy solid line of Fig. 1. A large conductance minimum centered around $V_{SD} = 0$ is observed accompanied by a series of conductance oscillations. We have observed similar conductance versus voltage curves for over ten similar samples, all yielding very dramatic conductance minima accompanied by a superimposed oscillatory feature.

Applying gate voltage, V_g, modifies these traces in a non-trivial way. The conductance versus gate voltage at low V_{SD} depicted in the top panel of Fig. 2 reveals pronounced conductance oscillations, which are attributed to the CB phenomena [12]. Fig. 2 shows a series of differential conductance versus V_{SD} curves taken for different gate voltages which span a typical CB oscillation.

The analysis of our findings is based on the theoretical treatment of a strongly coupled dot by Golubev *et. al.* [7] summarized in Eq. (1). The general expressions for the functions $I_0(T,V)$ and $F(T,V)$ are quite cumbersome and not very transparent [7]. However they can be considerably simplified for the experimentally relevant situation. For the low temperature limit ($T \ll \hbar/2\pi\ R_D C$) and asymmetrically coupled dot ($R_S \gg R_D$) one finds

$$I_0(T,V) = \frac{e^2}{2\pi\hbar}\frac{R_D}{R_S}V\log\left(1+\frac{\hbar^2}{4\pi^2\epsilon^2 t_c^2}\right)\ , \qquad (3)$$

where $\epsilon = \max\{eV, T\}$ and $t_c = R_D C$.
For the exponential factor $F(T,V)$ one finds

$$F(T,V) = \frac{\hbar}{2e^2 R_D} + \frac{2\pi^2}{e^2}CT + \frac{2\pi}{e^2}\sum_{r=D,S}\frac{1}{R_r}y(x_r). \quad (4)$$

Here $x_r = R_S R_D R_r eVC/(R_S+R_D)^2$, and $y(x) = x\arctan x - \frac{1}{2}\ln\left(1+x^2\right)$ and $C = C_S + C_D + Cg$.

Differentiating Eq. (1) utilizing the expressions derived in Eqs. (3) and (4) and assuming a very asymmetric dot at small voltages we reach the following expression for the differential conductance through the dot:

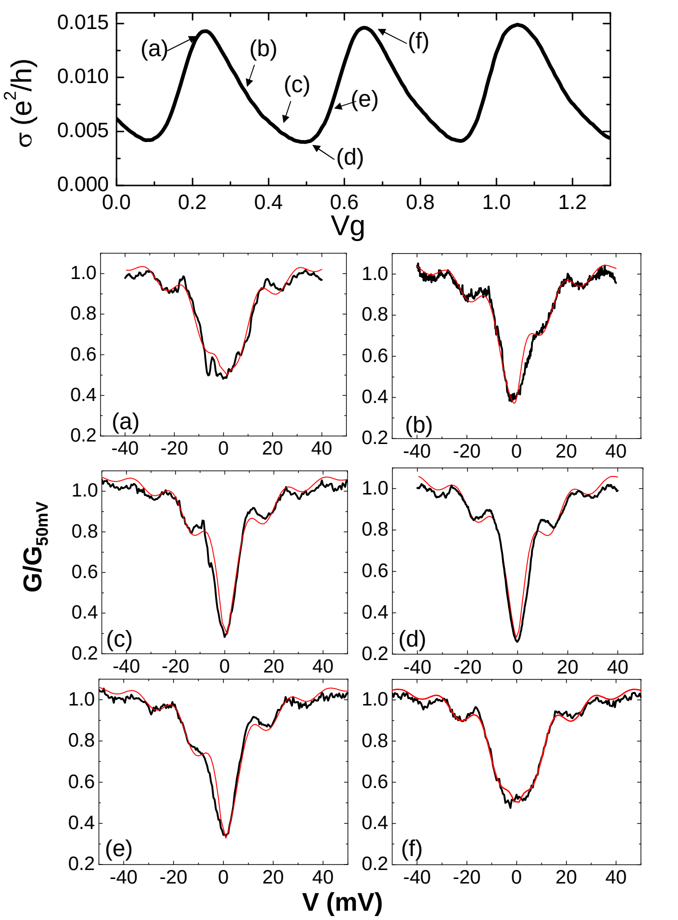

FIG. 2: Top: conductance versus gate voltage at low V_{SD} of a quantum dot system measured at T=4.2K exhibiting CB features. a-f: Differential conductance versus V_{SD} for a series of gate voltages corresponding to the marked points in the top figure. The light solid red lines are fits to Eq. (5).

$$\frac{G(V)}{G_{as}} = 1 - \frac{1}{g_D}\left(\ln\left[1+\frac{\hbar^2}{(2\pi t_c \epsilon)^2}\right] - \frac{2}{1+(2\pi t_c \epsilon/\hbar)^2}\right)$$
$$+\exp\left[-\frac{g_D}{2} - \frac{2\pi^2}{\tilde{E_C}}\left(T + \frac{R_D}{R_S}V_{SD}\right)\right]\cos\left(\frac{2\pi V_{SD}}{E_C}+\phi\right), \quad (5)$$

where $g_D = \frac{h}{e^2 R_D}$ is the dimensionless conductance between the dot and the drain (the well connected electrode), t_c is the charging time of the dot, $E_C = \frac{e^2}{C_S}$ is the charging energy determining the staircase period, $\tilde{E_C} = \frac{e^2}{C}$ determines the amplitude of the CB oscillations, $\phi = CgVg/e$ is the phase of the CS which is sensitive to the applied gate voltage and $\epsilon = (V^2 + \tilde{T}^2)^{1/2}$ is the energy of the system. To fit our data we notice that the smearing of ZBA is determined by an effective temperature $\tilde{T} = 2mV$, which is a factor of 5 larger than the experimental temperature and will be discussed later on.

Eq. (5) includes two distinct parts. The first term is a conductance dip centered at $V_{SD} = 0$ which we interpret as a 0D version of the ZBA for the well connected dot. As for the higher dimensional cases discussed by Altshuler and Aronov[1], the dip magnitude is inversely proportional to g and always pinned to the Fermi energy, i.e., centered at $V_{SD} = 0$. The second term is oscillatory with V_{SD}, with a period corresponding to the charging energy of the dot E_C. This term corresponds to the CB, it is sensitive to the gate voltage via the phase ϕ and is suppressed exponentially with g_D as predicted for the CB phenomena [5, 6] and with the V_{SD} and T. We emphasize that the same fitting parameters were used for all cases with only ϕ varying between the different curves a-f, thus "sliding" the CS along the voltage axis. These fits yield $g_D = 3.5$ or $R_D = 7.4k\Omega$, demonstrating that, contrary to the orthodox convention, CB effects can be measured even for $g > 1$. We note that we use the same g_D for both terms of Eq. (5) thus increasing our confidence in the fitting procedure.

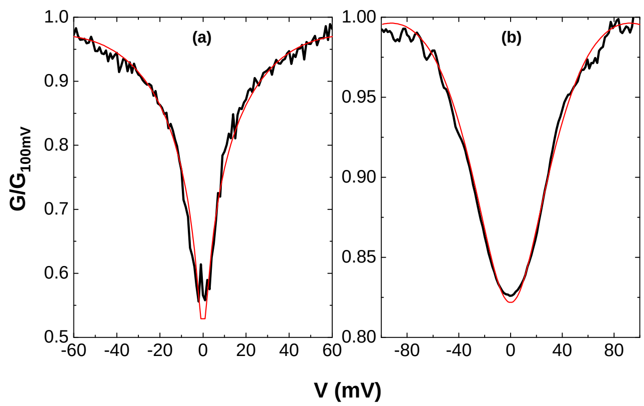

FIG. 3: (a) Conductance versus SD voltage for the case where the dot of Fig. 2 was coupled more strongly to the drain. The light solid red line is the fit to Eq. (5). (b) Conductance versus SD voltage for the dot of Fig 2. at $T = 77k$ with the fit to Eq. (5). In both cases only a dip around $V_{SD} = 0$ appears with no measurable superimposed oscillatory feature.

The CB term shows a difference of its suppression by temperature and source-drain voltage. While the temperature scale is determined by $\tilde{E_C}$, the voltage suppression is determined by $\frac{R_S}{R_D}\tilde{E_C} \gg \tilde{E_C}$. This explains why CB oscillations can be seen for $V_{SD} \sim 50mV$ while for $T = 77K \sim 8mV$ they are completely suppressed as seen in Fig. 3. The physical origin of the different behavior of the temperature and SD voltage stems from their different influence on inelastic processes of electrons in the dot. While temperature affects the occupation of all the electrons in the dot, the SD voltage influences only electrons tunneling in or out of it. Since the vast majority of electrons enter (or leave) the dot through the low resistance connection to the lead the relevant voltage scale is proportional to the voltage drop on it, i.e., to $\frac{R_S}{R_D}V_{SD}$.

As the coupling between the dot and the drain is increased all non-ohmic features in the I-V are sup-

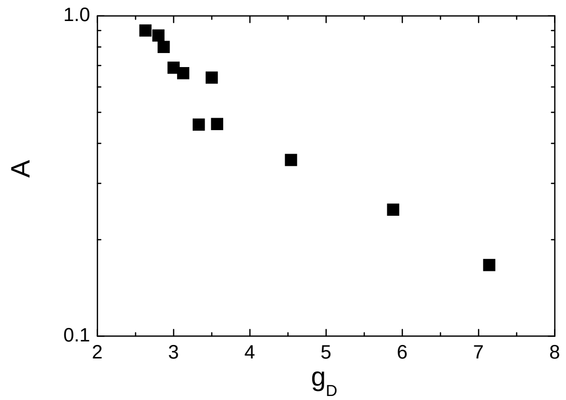

FIG. 4: Amplitude of the CB oscillations at small V_{SD} (such as those of Fig. 2a) defined as $[g_{max} - g_{min}]/g_{max}$ as a function of g_D extracted from the fits to Eq. (5) for 11 different dots.

pressed. However, the oscillatory feature is suppressed much faster. Fig. 3a shows the differential I-V curve for the same dot depicted in Fig. 2 in which the coupling to the drain has been increased. The I-V in this case exhibits only a ZBA like feature with no signs for CS effects. For this coupling the fit to Eq. (5) is achieved for $g_D = 5.5$. This result indicates that the CB effect is much more sensitive to coupling than the ZBA feature, as expected from Eq. (5).

Extracting g_D out of the ZBA feature in 11 measurements performed for different dots and comparing the amplitude of the CB oscillations of each case at $V_{SD} \approx 0$ (see Fig. 4) one can clearly see an exponential decrease of the CB amplitude as g_D increases which fits very well the explicit expression appearing in Eq. (5), i.e. the amplitude decreases as $\exp(-g_D)$. This fit provides an experimental verification of the common theoretical prediction that the CB amplitude should be suppressed exponentially with g in the regime $g \geq 1$ and also reinforces the confidence in the consistency of the analysis.

A similar suppression is seen when the temperature is increased. Fig. 3b shows the I-V characteristics through the dot at $T = 77K$. It is seen that at this temperature only a ZBA-like feature is observed and the CS has vanished. Indeed, CB effects are expected to decay exponentially with temperature (see Eq. (5)). Nevertheless an effective temperature of $\tilde{T} = 27mV$ in the ZBA term (more than 4 times the actual temperature) is needed to achieve a good fit (light solid red line).

Thus the experimental results seem to be well described by our approximation of the theory of Golubev *et. al.* [7] depicted in Eq. (5). There remains though the puzzle of the relative high effective temperature $\tilde{T}$ needed for the fit of the ZBA term in Eq. (5). $\tilde{T}$ corresponds to smearing of the ZBA dip. The fact that the smearing of the dip is stronger than expected from the system's temperature indicates that an additional physical mechanism contributes to inelastic processes in the dot. In contrary to typical quantum dot systems the grain here is mechanically free standing, although there is a good electrical contact. Mechanical vibrations, which might induce fluctuations in the tunneling to the dot might significantly contribute to the dephasing and inelastic processes manifested in the smearing of the ZBA.

In conclusion, we have investigated the I-V characteristics of transport through an asymmetrically coupled metallic grain, which is well connected to one of the leads, but poorly connected to the other. This is an unusual regime for which properties usually associated with weakly coupled 0D systems (CB staircases) and disordered higher dimensional leads (Altshuler-Ahronov ZBA) appear and may be easily separated. Identifying the relevant limits in the general expression of Golubev *et. al.* we have shown that in our case the CB and ZBA are both relevant and show a different dependence on the strong coupling of the grain to the lead (g_D), temperature, gate and source-drain voltage. We use these features to determine the parameters of the transport (such as the source and drain resistances) which can not be separated in regular transport measurements.

We are grateful for useful discussions with Y. Gefen and A.D. Mirlin and especially insightful input from I.V. Gornyi. This research was supported by the Israeli academy of science (grant number 399/09)

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